

## LETTER TO THE EDITOR

## Breit interaction in heavy atoms

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**Abstract.** In this letter we discuss how to include Breit interaction in calculations of low-energy properties of heavy atoms in accurate and effective way. In order to illustrate our approach we give some results for Cs and Tl.

*Introduction.* It is well known that in high accuracy atomic calculations one may need to go beyond the Coulomb approximation for the two-electron interaction. The usual next step is to include the Breit interaction. A number of commonly used atomic packages, such as GRASP package [1], allows to do this routinely. However, in many precision calculations one needs to account for different corrections, which are not included in standard packages. Thus, it may be important to analyze the role of the Breit interaction on different stages of atomic calculations. Below we present several simple observations which may help to include Breit interaction in atomic calculations in an optimal way.

Breit interaction between electrons 1 and 2 has the form

$$V_B \equiv V_B^1 + V_B^2 = -\frac{\vec{\alpha}_1 \cdot \vec{\alpha}_2}{r_{12}} + \frac{1}{2} \left\{ \frac{\vec{\alpha}_1 \cdot \vec{\alpha}_2}{r_{12}} - \frac{(\vec{r}_{12} \cdot \vec{\alpha}_1)(\vec{r}_{12} \cdot \vec{\alpha}_2)}{r_{12}^3} \right\}, \quad (1)$$

where  $\vec{\alpha}_i$  are Dirac matrices and  $r_{12}$  is the distance between the electrons. Equation (1) can be readily derived from the general expression for the interaction via transverse photon in the Coulomb gauge by expanding it in powers of the photon frequency (see, for example, [2]). The operator  $V_B^1$  is called the magnetic (or Gaunt) term and the operator  $V_B^2$  is called the retardation term [3]. The magnetic term of the Breit operator is known to dominate in atomic calculations [4] and it is much simpler than the whole Breit operator. In present work we will neglect the retardation term and we will check the accuracy of this approximation by comparison to the resent calculation by Derevianko [5] where the whole Breit operator was used. Note also, that some technical details of atomic calculations with retardation term were recently discussed in [6] (see also references therein).

*Breit interactions in Dirac-Fock calculations.* Here, we are interested in the calculations of the low-energy properties of heavy atoms. In such calculations all electrons are usually divided into two parts: the core electrons and the valence ones. Correspondingly, the interaction between electrons is reduced to the interaction between the valence electrons and the interaction of the valence electrons with atomic core. The latter is described by the Hartree-Fock potential which includes direct and exchange terms. In this case the following observations can be made.

- (i) Breit correction to the interaction between the valence electrons is of the order of  $\alpha^2$  ( $\alpha$  is the fine structure constant) which is usually below the accuracy of modern calculations of heavy atoms.
- (ii) Breit correction to the direct term of the Hartree-Fock potential turns to zero if the core includes only closed shells. Indeed, the vertex of the Breit interaction includes Dirac matrix  $\vec{\alpha}$ , which is averaged to zero when summation over the closed shell is done.
- (iii) Breit correction to the exchange interaction of the valence electron with the core does not turn to zero and is of the order of  $R^2$ , where  $R$  is the overlap integral between the upper component of a valence orbital and the lower component of a core one. The largest integrals  $R$  correspond to the innermost core orbitals, where small components are of the order of  $\alpha Z$ . Thus, the dominant Breit correction is the one to the exchange core potential.
- (iv) The exchange interaction between valence electrons and the innermost core electrons is significantly screened if the core relaxation is allowed. Therefore, it is very important that Breit correction to the core potential is calculated self-consistently. In some cases the core relaxation can reduce the final Breit correction to the valence energies by an order of magnitude (see the results for Cs below).

We are not going to give all technical details of the calculations here, but there are few points to be mentioned, at least briefly. Above we have argued that the only important contribution of the Breit interaction comes from the exchange with the core. The corresponding matrix element can be written in a form:

$$\begin{aligned} \langle f | V_B^{\text{core}} | i \rangle &= - \sum_{c \in \text{core}} \langle f, c | V_B | c, i \rangle \\ &= -\delta_{j_f, j_i} \delta_{m_f, m_i} \sum_k \sum_{c \in \text{core}} (2j_c + 1) \begin{pmatrix} j_f & j_c & k \\ -\frac{1}{2} & \frac{1}{2} & 0 \end{pmatrix}^2 R_{f, c, c, i}^k, \end{aligned} \quad (2)$$

where  $R_{f, c, c, i}^k$  is the radial integral of the Breit interaction for multipolarity  $k$ .

Interaction (2) can be included in calculation either perturbatively or self-consistently. In the former approach, the first order correction to the energy of the valence electron  $v$  is simply  $\delta\varepsilon_v = \langle v | V_B^{\text{core}} | v \rangle$ . In the self-consistent approach, the potential  $V_B^{\text{core}}$  should be included in the Dirac-Fock (DF) equations. These equations will then give new set of orbitals and energies  $\{\tilde{\varphi}_n, \tilde{\varepsilon}_n\}$  and  $\delta\varepsilon_v = \tilde{\varepsilon}_v - \varepsilon_v$ .

There are two things one has to keep in mind when solving the Dirac-Fock-Coulomb-Breit (DFCB) equations. The first is that Breit approximation is not completely relativistic and, thus, some caution may be necessary. For example, one can use projectors to the positive energy states. Technically that can be done with the help of the kinetic balance condition for the small components of the Dirac orbitals. Second, if we include Breit interaction in DF equations, the resultant corrections are not linear in Breit interaction. It is not difficult to eliminate the higher orders in Breit interaction with the help of a scaling parameter  $\lambda$ : one can substitute  $V_B$  with  $\lambda V_B$  and then calculate  $\lim_{\lambda \rightarrow 0}(\delta\varepsilon_n^\lambda/\lambda)$  and  $\lim_{\lambda \rightarrow 0}(\delta\varphi_n^\lambda/\lambda)$ . In practice, however, the higher orders in Breit interaction are usually small enough to be neglected even for  $\lambda = 1$  and there is no need in calculating these limits.

*Correlation effects.* The usual accuracy of the DF approximation for binding energies of heavy atoms is of the order of 10%, while the Breit corrections are usually about 1% or less. Therefore, there is no point in calculating Breit corrections if one is not going to account for correlations in some way. It is usually convenient to distinguish between valence-valence and valence-core, or core-core correlations. The easiest and straightforward way to treat the former is to use the configuration interaction (CI) method, while the latter are treated more efficiently within the many-body perturbation theory (MBPT) in residual Coulomb interaction [7]. Below we discuss how to include Breit interaction in the CI and the MBPT calculations.

If the CI is done in the frozen core approximation, then as we have said above, there is no need to include Breit corrections to the two-electron interaction, but it is necessary to include Breit corrections to the core potential. It is also important that core orbitals are found from the DFCB equations.

If the MBPT in residual Coulomb interaction is used to account for valence-core and core-core correlations, one may need to include Breit corrections to corresponding diagrams. Generally speaking, there are two types of corrections: “direct” and “indirect” ones. The former corresponds to the substitution of the residual Coulomb interaction with the Breit interaction in the MBPT expressions, and the latter corresponds to the use of the DFCB equations as a zero order approximation. The direct corrections are suppressed because the largest Breit radial integrals correspond to the virtual excitations from the innermost core shells (see above) and these excitations correspond to the huge energy denominators. Therefore, one can neglect them without significant loss of accuracy. The indirect corrections are much simpler: they are accounted for simply by the use of the DFCB basis set instead of the DFC one.

We see that dominant Breit corrections come from the solution of the DFCB equations for atomic orbitals. After that these orbitals can be used as a basis set for the CI and the MBPT calculations. On the CI stage one has to include Breit potential of the core explicitly when calculating one-electron integrals for valence electrons, while in the MBPT calculations the direct Breit corrections can be neglected altogether. That significantly simplifies calculations and allows to use standard CI and MBPT codes

without significant changes.

Up to now we have focused on the calculations of atomic spectra. If it is necessary to calculate some other atomic observables, the general calculation scheme remains the same [8]. However, the MBPT part should be extended and some new types of the MBPT corrections may appear. Also, it may be necessary to solve the random-phase approximation (RPA) equations for the operator of interest. For example, if we calculate hyperfine structure (HFS) constants, we have to solve the RPA equations for the operator of the hyperfine interaction. Again there are direct and indirect Breit corrections to these equations. The latter are easily included in the same way as above. The former are important only for operators which are singular at the origin and, thus, have large matrix elements for the innermost core electrons. This is the case for the hyperfine interaction, while electric dipole operator gives the opposite example.

*Numerical results.* Here we present some results for Cs and Tl. These two atoms are interesting because they are used in precision measurements of parity nonconservation and high accuracy atomic calculations are necessary to compare these measurements with Standard model predictions. The nuclear charge  $Z$  for Tl is much larger than for Cs, so we can also see how Breit corrections grow with the nuclear charge.

In table 1 we present results of the DF calculations of binding energies for Cs in the Coulomb and Coulomb-Breit approximations. We also give results of the perturbative calculations of the Breit corrections for comparison. It is seen that the relaxation of the core significantly reduces Breit corrections to the valence energies.

In table 2 the direct Breit corrections to the HFS constants are given. As we have mentioned above, here one has to calculate Breit correction to the RPA vertices in addition to the corrections to the orbitals. In notations of Derevianko [5] the latter corresponds to the one-particle correction and the former corresponds to the two-particle one. It is seen again that corrections obtained in the self-consistent approach are much smaller than the corresponding first order perturbative corrections. That is caused by the screening of the Breit interaction by the core electrons which is neglected in the perturbative approach. Note, that screening effect is particularly important for the  $s$ -wave.

The difference between our calculations of the first order Breit corrections to the HFS constants and the results of the paper [5] should be due to the retardation term in (1). This difference is about 10% for the  $p$ -wave and 25% for the  $s$ -wave. That confirms that this term is significantly smaller than the magnetic one, but it can become important when Breit correction becomes larger or the accuracy of the calculations is improved.

On the next stage we calculated correlation corrections following the method described in [7, 8]. These corrections include the self-energy correction to the Hartree-Fock potential and the structural radiation corrections to the HFS operator. The latter contribution is small and we did not calculate Breit corrections to it. Note that the indirect Breit corrections were calculated for the self-energy and the RPA contributions,

while the direct Breit corrections for the self-energy were neglected. The results of these calculations are given in table 3. It is seen that direct and indirect Breit corrections to the HFS constants are comparable. That means that it is important to include Breit corrections when electronic correlations are considered.

Calculations for Tl are much more complicated and require large CI for three valence electrons in addition to the MBPT (see [7, 8] for details). The Breit corrections appear to be much more important for Tl than for Cs. Here they are not negligible already when the energy spectrum is considered (see table 4). In particular, the fine structure splitting of the ground state is changed by 1%. Another difference from Cs is that the screening effect here is relatively weaker, but still significant. For example, the first order Breit correction to the fine splitting of the ground state is  $-130 \text{ cm}^{-1}$  while the DFCB value is  $-81 \text{ cm}^{-1}$ .

Comparison of the results of the CI calculations from table 4 with the final results, which include the MBPT corrections, demonstrates that Breit contributions to the MBPT part constitute about 10% of the total Breit corrections to the transition frequencies. That is consistent with the overall role of the MBPT corrections in the final answer where they also contribute about 10%.

*Conclusions.* We see that Breit interaction is important in precision calculations of heavy atoms. It is sufficient to include only (exchange) Breit potential of the core and neglect valence-valence Breit interaction. It is important to allow core relaxation to avoid overestimation of Breit corrections to energies and orbitals of valence electrons. It is also important to calculate Breit corrections to the RPA part and to the MBPT part when one calculates the HFS constants or other similar observables.

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**Table 1.** Binding energies for Cs (au) in Dirac-Fock-Coulomb (DFC) and Dirac-Fock-Coulomb-Breit (DFCB) approximations. The last row gives the one-particle first order Breit (FOB) correction in the perturbative approach.

	$6s_{1/2}$	$6p_{1/2}$	$6p_{3/2}$	$7s_{1/2}$	$7p_{1/2}$	$7p_{3/2}$
DFC	0.127368	0.085616	0.083785	0.055187	0.042021	0.041368
DFCB	0.127358	0.085577	0.083768	0.055183	0.042008	0.041362
FOB	0.127217	0.085537	0.083726	0.055146	0.041993	0.041347

**Table 2.** The direct Breit corrections to the hyperfine structure constants in Cs(MHz). Corrections to the orbitals are calculated perturbatively (FOB) and self-consistently (DFCB). Direct corrections to the RPA are calculated as discussed in the text.

	DFC	FOB		DFCB	RPA	
		this work	[5]		this work	[5]
$A_{6s_{1/2}}$	1423.8	-10.7	-8.14	-1.2	3.1	3.50
$A_{6p_{1/2}}$	160.9	-1.9	-1.58	-1.1	0.7	0.73
$A_{6p_{3/2}}$	23.9	-0.2	—	-0.1	0.1	—
$A_{7s_{1/2}}$	391.2	-2.4	-1.80	0.0	0.9	0.96
$A_{7p_{1/2}}$	57.6	-0.6	-0.54	-0.4	0.2	0.26
$A_{7p_{3/2}}$	8.6	0.0	—	0.0	0.0	—

**Table 3.** Final results with the MBPT corrections for the HFS constants for Cs (MHz) in Coulomb and Coulomb-Breit approximations. The definitions of the direct Breit (DB) and the indirect Breit (IB) corrections are given in the text. Experimental result are taken from [9, 10].

	Coulomb	DB	IB	Total	Exper.
$A_{6s_{1/2}}$	2312	1.9	0.9	2315	2298
$A_{6p_{1/2}}$	296	-0.4	-0.4	295	292
$A_{6p_{3/2}}$	55.3	0.0	-0.1	55.2	50.3
$A_{7s_{1/2}}$	549	0.9	-0.9	549	546
$A_{7p_{1/2}}$	94.0	-0.2	-0.1	93.6	94.3
$A_{7p_{3/2}}$	18.2	0.0	0.0	18.2	18.2

**Table 4.** Calculation of the spectrum of Tl in the Coulomb (C) and the Coulomb-Breit (CB) approximations (in  $\text{cm}^{-1}$ ). The DF equations were solved in the  $V^{N-1}$  approximation. The CI was done for three valence electrons and final calculation included core-valence and core-core correlations within the CI+MBPT method.

	DF		CI		Final		Exper. [11]
	C	CB	C	CB	C	CB	
$6p_{1/2}$	0	0	0	0	0	0	0
$6p_{3/2}$	7186	7105	7066	6988	7878	7780	7793
$7s_{1/2}$	22713	22597	24767	24660	26596	26474	26478
$7p_{1/2}$	29546	29434	32210	32108	34109	33993	34160
$7p_{3/2}$	30465	30346	33159	33048	35119	34994	35161
$6d_{3/2}$			33970	33851	36175	36041	36118
$6d_{5/2}$			34029	33909	36250	36115	36200